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CLAIMS

[Claim(s)]

[Claim 1] The manufacturing installation of the fullerene which has the decollator which separates the fullerene of a gaseous state, and/or the multi-annular aromatic compound of a gaseous state from the air current which contains fullerene, a multi-annular aromatic compound and the fullerene generation equipment that makes the air current containing the soot-like matter containing a carbon system macromolecule component generate, and the soot-like matter from a carbonaceous raw material.

[Claim 2] Furthermore, the manufacturing installation of the fullerene according to claim 1 characterized by having a decompression device for decompressing generation equipment and a decollator, and connecting fullerene generation equipment, the decollator, and the decompression device to this order.

[Claim 3] Furthermore, it is the manufacturing installation of the fullerene according to claim 1 or 2 to which it has deposit equipment which deposits the fullerene of a gaseous state which passed the decollator, fullerene generation equipment, a decollator, and deposit equipment are connected to this order, and a decompression device is characterized by preparing the front stirrup of deposit equipment behind.

[Claim 4] The manufacturing installation of the fullerene according to claim 1 to 3 further characterized by preparing the isolation region which separates fullerene according to molecular weight into deposit equipment.

[Claim 5] The manufacturing installation of the fullerene according to claim 1 to 4 which has the filter with which a decollator passes the fullerene of the gaseous state included in the soot-like matter, and/or the multi-annular aromatic compound of a gaseous state, and is characterized by the heat-resistant temperature of this filter being 300 degrees C or more.

[Claim 6] the combustion method to which the generation method of the fullerene in fullerene generation equipment carries out the incomplete combustion of the hydrocarbon raw material - or the manufacturing installation of the fullerene according to claim 1 to 5 characterized by being the thermal decomposition method which pyrolyzes a hydrocarbon raw material.

[Claim 7] The manufacturing installation of the fullerene according to claim 1 to 6 to which a decollator is characterized by having a recovery means to collect the carbon system macromolecule components separated from the soot-like matter content air current.

[Claim 8] The manufacture approach of the fullerene which has the following processes (1) and processes (2).

Process (1): The process which makes the soot-like matter content air current which contains fullerene, a multi-annular aromatic compound, and a carbon system macromolecule component by carrying out the incomplete combustion of the hydrocarbon raw material, or pyrolyzing a hydrocarbon raw material generate.

Process (2): The process which separates fullerene and/or a multi-annular aromatic compound from the fullerene and the multi-annular aromatic compound which were obtained at the process (1), and the soot-like matter content air current containing a carbon system macromolecule component by the gaseous state.

[Claim 9] The manufacture approach of the fullerene according to claim 8 characterized by

separating fullerene from the soot-like matter containing the fullerene and the carbon system macromolecule component which separated the multi-annular aromatic compound by the gaseous state in the process (2), and were subsequently obtained.

[Claim 10] The manufacture approach according to claim 8 or 9 characterized by using the 400-1500-degree C soot-like matter content air current acquired at the process (1) at a process (2).

[Claim 11] The manufacture approach of the fullerene according to claim 9 or 10 characterized by having the process which extracts fullerene with the solvent containing an aromatic hydrocarbon compound, and is separated from the soot-like matter containing the fullerene and the carbon system macromolecule component from which the multi-annular aromatic compound might be separated at the process (2).

[Claim 12] The manufacture approach of the fullerene according to claim 9 or 10 characterized by having the process which separates fullerene by the gaseous state from the soot-like matter content air current containing the fullerene and the carbon system macromolecule component from which the multi-annular aromatic compound might be separated at the process (2).

[Claim 13] The manufacture approach of the fullerene according to claim 8 which cools, uses as such shape of a solid-state and liquid mixture the fullerene and the multi-annular aromatic compound which were obtained by the gaseous state through the process (2), and is characterized by having the process which the solubility of fullerene extracts a multi-annular aromatic compound, and separates from this mixture with an organic solvent with the high solubility of a multi-annular aromatic compound low.

[Claim 14] The manufacture approach of the fullerene according to claim 8 which cools, uses as such shape of a solid-state and liquid mixture the fullerene and the multi-annular aromatic compound which were obtained by the gaseous state through the process (2), and is characterized by having the process which heats this mixture and separates a multi-annular aromatic compound by the gaseous state.

[Claim 15] The manufacture approach of the fullerene according to claim 8 characterized by having the process which separates fullerene as a solid-state, cooling the fullerene and the multi-annular aromatic compound which were obtained by the gaseous state through the process (2), and making a multi-annular aromatic compound into a gaseous state.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacturing installation and the manufacture approach of the fullerene which is a new carbon material, and the fullerene which has the molecular structure of C60, C70, C76, C78, C82, and C84 especially.

[0002]

[Description of the Prior Art] The molecule-like carbon matter new type called the carbon cluster (spherical macromolecule) of the closed shell structured type of carbon numbers 60 and 70 and 84 grades is compounded, and attracts attention in 1990. The carbon cluster which has this special molecular structure is also called fullerene, and is called fullerene C60, this C70, this C84, etc. by the carbon number which constitutes that molecule frame (only called C60, C70, and C84 grade). These fullerene (it may only be hereafter called "fullerene") is new carbon materials, and since it is expected that unique physical properties will be shown from the special molecular structure, research on the property and application development is advanced briskly. As for fullerene, use in fields, such as for example, diamond coating, a cell ingredient, a coating, a heat insulator, lubricant, drugs, and cosmetics, is expected. In the field of (1) - (4) specifically shown below, innovative application development is being developed quickly.

[0003] (1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.

(2) Application to drugs : by using C60 derivative etc., research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis, the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced.

(3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.

(4) Application to semi-conductor manufacture : by mixing C60 with a photopolymer (resist), it uses that resist structure is strengthened further and the application to next-generation semi-conductor manufacture is expected.

[0004] As the manufacture approach of fullerene, the electrode which consists of carbonaceous ingredients, such as (1) graphite, is used as a raw material. By the pulse laser exposure of the approach (arc discharge method) of evaporating a raw material by making inter-electrode [this] producing arc discharge, the method (resistance heating method) of passing a high current in (2) carbonaceous raw materials, and evaporating a raw material, and (3) high-energy consistency The approach (combustion method) of carrying out the incomplete combustion of the organic substance, such as a method (laser evaporation method) of evaporating a carbonaceous raw material and (4) benzene, etc. is learned.

[0005] however, the present condition -- two or more fullerene which cannot manufacture only the target single fullerene or fullerene of useful C60 - useful C84 grade, but is mainly concerned with C60 and C70 by any manufacture approach -- in addition, mixture (this products of combustion may be called the "soot-like matter") with many carbon compounds will be

generated. Since it is about 10 – 30% also by the arc discharge method said for the content of the fullerene in this soot-like matter to be efficient, in order to obtain the fullerene of a high grade, it is necessary to separate fullerene from the soot-like matter.

[0006] As the separation approach of the fullerene from the soot-like matter, the solvent extraction method is known, for example. The carbon system giant-molecule component which occupies many of other soot-like matter to fullerene dissolving this in organic solvents, such as benzene, toluene, and a carbon disulfide, and which is the so-called "soot" is an approach using the property to be hard to dissolve in an organic solvent like graphite or amorphous carbon. Moreover, as an option which separates fullerene from the soot-like matter, the soot-like matter is heated under a high vacuum, and the approach (the sublimating method) to which fullerene is made to sublimate is learned. This sublimating method is the special separation approach which needs an elevated temperature 400 degrees C or more and the high vacuum conditions below 0.133Pa (10⁻³Torr), and since a solvent extraction method is easy to operate it compared with it, it is used widely. The fullerene furthermore obtained by the extract is a solution which mainly contains that of C60 and C70, and in order to separate single fullerene from this solution, the approach of carrying out clathration of the fullerene else [, such as column chromatography separation and judgment recrystallization,] etc. is applied.

[0007] In addition, after carrying out contact processing of the solution which extracted the soot-like matter containing C60 with the organic solvent as the fullerene separation approach from the soot-like matter, and was obtained and which mainly contains C60 with activated carbon, there is a method of refining C60 by carrying out separation removal of the organic solvent from the obtained processing liquid (for example, refer to patent reference 1.).

Moreover, as a separation refiner of fullerene, the separation refiner of the small quantity and batch type containing a heating container, a trap, and a decompression device is known. It specifically connects with the heating container and this heating container for making fullerene heat and sublimate, and has a decompression device for carrying out reduced pressure suction of the interior of the trap for depositing the sublimated fullerene, and a heating container and a trap at least, and the heating container, the trap, and the decompression device are arranged in this sequence (for example, refer to patent reference 2.).

[0008]

[Patent reference 1] JP,5-85711,A [the patent reference 2] JP,9-227111,A [the patent reference 3] U.S. Pat. No. 5273729 specification [0009]

[Problem(s) to be Solved by the Invention] However, each of these separation purification approaches is batch types, and is the purification techniques for the little soot-like matter. Therefore, it is unsuitable for fullerene manufacture on the industrial scale which makes a lot of soot-like matter applicable to purification. Moreover, current [which is represented by carbon black], the carbon system macromolecule component which is worthy soot industrially are contained in this soot-like matter in large quantities. Therefore, from a lot of soot-like matter, also in order to also use these carbon system macromolecule component effectively, development of the manufacturing installation of fullerene which can separate a carbon system macromolecule component and fullerene as efficiently as possible, and the manufacture approach was desired.

[0010] Moreover, also in the manufacture approach of fullerene, the multi-annular aromatic compound generated with fullerene had become a problem. Although the soot-like matter containing two or more fullerene which is mainly concerned with C60 and C70 generates when manufacturing fullerene by the approach (combustion method) of carrying out the incomplete combustion of the organic substance, such as toluene, under the temperature and the flow and pressure requirement specifically controlled, it is known that 10 ppm – 5% of the weight of the multi-annular aromatic compound is contained in this soot-like matter (for example, refer to patent reference 3.).

[0011] Also in an aromatic compound, in presentation, as for this multi-annular aromatic compound, the appearance represented by the benzopyrene has few rates of a hydrogen atom, and it is similar to it with fullerene. Therefore, when intermingled in fullerene, the reactivity of

fullerene may be checked, or the property of the proper of fullerene may be affected. Moreover, a these multi-annular aromatic compound is poison, and it is necessary to reduce it as much as possible from the field of safety generally.

[0012] However, when the solubility to the solvent of fullerene and a multi-annular aromatic compound is measured, generally the solubility of a multi-annular aromatic compound is higher 10 or more times. If a solvent extracts the soot-like matter for the reason, it will be difficult to extract fullerene alternatively and it will extract most multi-annular aromatic compounds in the soot-like matter to coincidence to an extract. Even if for that reason it condenses and dries an extract, or it carries out solid content which condensed the extract and deposited a ** exception, dries and it obtains a solid-state (fullerene is mainly included) in order to obtain fullerene from this extract as a solid-state, the problem that about 0.01 – 10% of multi-annular aromatic compound will usually be contained is in this solid-state. This invention is made in view of a situation which was mentioned above, and it aims at offering the manufacturing installation and the manufacture approach of carrying out separation recovery of the fullerene efficiently from a lot of soot-like matter to generate in the case of extensive manufacture of fullerene.

[0013]

[Means for Solving the Problem] Generally, once a lot of existing carbon system macromolecule components in the soot-like matter become a solid-state, it is known to not changing to a gaseous state easily that fullerene will change to a gaseous state in a temperature field 400 degrees C or more.

[0014] The sublimation temperature of fullerene paid its attention to this invention persons that it is low temperature comparatively of 400 degrees C or more. And the soot-like matter was obtained as the condition which floated in the air current in the fullerene manufacture approach, and a specific combustion method and a specific thermal decomposition method specifically using the carbonaceous raw material, i.e., a soot-like matter content air current, and since this air current was usually the elevated temperature of existing extent, fullerene existed by the gaseous state, and I thought that a carbon system macromolecule component was in a solid state.

[0015] And as a result of this invention persons' inquiring wholeheartedly, from the soot-like matter content air current acquired by such specific approach, it found out that fullerene was separable from the soot-like matter containing the carbon system macromolecule component which also produces ** in large quantities in the manufacture process of fullerene not using an organic solvent and which is the so-called soot by the gaseous state. Furthermore, artificers found out that it was also disengageable at a gaseous state also in the serious harmful multi-annular aromatic compound contained in this soot-like matter. That is, since the sublimation temperature of this multi-annular aromatic compound is lower than that of fullerene The fullerene, the multi-annular aromatic compound which were obtained in the combustion method or the thermal decomposition method. And by heating the soot-like matter content air current containing a carbon system macromolecule component to the temperature which a multi-annular aromatic compound sublimates first, separating a multi-annular aromatic compound by the gaseous state, and subsequently heating a soot-like matter content air current further etc. A header and this invention were completed for fullerene being separable by the gaseous state.

[0016] That is, the summary of this invention consists in the manufacturing installation of the fullerene which has the decollator which separates the fullerene of a gaseous state, and/or the multi-annular aromatic compound of a gaseous state from the air current which contains fullerene, a multi-annular aromatic compound and the fullerene generation equipment that makes the air current containing the soot-like matter containing a carbon system macromolecule component generate, and the soot-like matter from a carbonaceous raw material.

[0017] Moreover, the unsatisfactory summary of this invention consists in the manufacture approach of the fullerene which has the following processes (1) and processes (2).

Process (1): The process which makes the soot-like matter content air current which contains fullerene, multi-annular aromatic compound fullerene, multi-annular aromatic hydrocarbon, and a

carbon system macromolecule component by carrying out the incomplete combustion of the hydrocarbon raw material, or pyrolyzing a hydrocarbon raw material generate.

Process (2): The process which separates fullerene and/or a multi-annular aromatic compound from the fullerene and the multi-annular aromatic compound which were obtained at the process (1), and the soot-like matter content air current containing a carbon system macromolecule component by the gaseous state.

[0018]

[Embodiment of the Invention] First, the manufacturing installation of fullerene is explained.

[0019] Drawing 1 is the whole example schematic diagram of the fullerene manufacturing installation concerning this invention at the time of using a combustion method in fullerene generation equipment. The fullerene manufacturing installation concerning this invention has the component of fullerene generation equipment 1, fullerene, and others, and the decollator 3 which specifically separates a multi-annular aromatic compound and a carbon system macromolecule component. It has preferably the deposit equipment 5 which deposits fullerene, and a decompression device 4 further. When using a combustion method for generation of fullerene, fullerene generation equipment 1 is generation equipment 1 which inert gas, such as helium, was made full and made the internal pressure reduced pressure conditions and the desirable condition near a vacuum to atmospheric pressure. Fullerene coal-for-coke-making-ized hydrogen, such as benzene introduced into the side face of this equipment from introductory tubing (not shown), is introduced in fullerene generation equipment 1 with oxygen. Moreover, in fullerene generation equipment 1, it has the burner 2 for making incomplete combustion cause coal-for-coke-making-ized hydrogen at least. The air current which contains the soot-like matter containing fullerene continuously according to such structure can be manufactured. By this invention, a lot of fullerene can be manufactured in succession by separating fullerene by the gaseous state from this soot-like matter content air current.

[0020] The decollator 3 of fullerene is installed in the interior or the lower stream of a river of fullerene generation equipment 1. Moreover, since what the inside of fullerene generation equipment 1 and a decollator 3 is generally made into the reduced pressure condition for is desirable in order to separate generation of fullerene, and fullerene by the gaseous state, it is desirable to have the decompression device 4 for decompressing the internal pressure of the generation equipment 1 of fullerene and a decollator 3. Although the soot-like matter content air current which contains fullerene from the generation equipment of fullerene flows with a decollator 3 and deposit equipment 5 in the manufacturing installation of the fullerene of this invention, since the flow of this air current is generally loose, in order to obtain fullerene efficiently, it is desirable to arrange a decompression device 4 following deposit equipment 5, and to pass an air current efficiently.

[0021] What has a well-known filter operation conventionally as a separation means to constitute a decollator 3 can be used. Usually, since generation of fullerene is performed under high temperature conditions, a heat-resistant filter is used as a filter used for the decollator 2 formed in the next of the generation equipment 1 of such fullerene. A heat-resistant filter is installed for the purpose for catching the soot component which exists as a solid-state at the temperature of 400 degrees C or more, and it is important for fullerene to install in the temperature field which sublimates and exists by the gaseous state. As such a filter, for example, the sintered metallic filter by the Japanese pole company, the sintered metallic filter by the Fuji filter company, etc. are mentioned. What is necessary is to choose the magnitude of a filter opening suitably and for the combustion conditions which make fullerene generate, and the description of the soot-like matter just to determine it. Since the sublimation temperature of fullerene changes also with degree of vacuums, it is important for it to install this filter in the location which can pass fullerene most efficiently with the degree of vacuum in a container.

[0022] As long as the quality of the material of this heat-resistant filter can bear an elevated temperature 300 degrees C or more, it may be anything, and a ceramic, a sintered metal, etc. are used suitably. Moreover, it is desirable that the carbon system macromolecule component by which uptake is carried out with this filter is equipped with the recovery means 6, such as

back wash equipment discharged continuously. The carbon system giant-molecule component obtained in this phase is effectively utilizable as coloring agents, such as ink and a coating, the carbon black for tires, or a fuel. If the material which constitutes fullerene generation equipment 1 and ***** 3 can bear above-mentioned temperature and a flow and pressure requirement, as the quality of the material, metals, such as quartz glass and stainless steel, the ceramics, glass, etc. will be mentioned, for example.

[0023] Moreover, the thing of arbitration can be used for the separation approach in a decollator 3 for a batch type, a fixed-bed mold, a fluid bed mold, a continuation mold, etc. And in a decollator 3, inert gas may be further added in the air current introduced from fullerene generation equipment 1. What is necessary is to prepare the input and the exhaust port of inert gas in the decollator, and just to make inert gas flow and discharge continuously or intermittently in that case. Moreover, the temperature of the inert gas made to flow into a decollator 3 may be adjusted, and the temperature in a decollator 3 may be risen or dropped to predetermined temperature. Although this decollator 3 may separate fullerene and a multi-annular aromatic compound into coincidence by the gaseous state, the same decollator for separating a multi-annular aromatic compound as a decollator 3 may be formed in front of this decollator 3. Under the present circumstances, a multi-annular aromatic compound and fullerene become disengageable by the gaseous state separately.

[0024] The fullerene which passed the decollator 3 is the so-called "**** fullerene" containing the high order fullerene which has C60, C70, and the molecular weight beyond this. What is necessary is just to use the difference of such sublimation temperature, in order to generate this according to fullerene molecular weight further. Since sublimation temperature differs, fullerene, such as C60 and C70, can carry out separation uptake of C60 and C70 by installing an isolation region (not shown) in the location of temperature where each will be from a gaseous state in a solid state, respectively. As for such an isolation region that separates fullerene according to molecular weight, preparing in deposit equipment 5 is desirable. As an isolation region, the same filter as the decollator 3 of point ** and the temperature in the deposit field 5 may be lowered gradually, and each fullerene may make the internal surface of deposit equipment 5 etc. carry out a sequential deposit. Uptake of the fullerene can be carried out by installing a filter etc. in the location which finally becomes the temperature below the sublimation temperature of fullerene.

[0025] Then, the manufacture approach of the fullerene of this invention is described.
Process (1)

In the process (1) of this invention, the soot-like matter content air current containing fullerene is acquired. Under the present circumstances, fullerene is manufactured with the combustion method to which the incomplete combustion of the hydrocarbon (coal-for-coke-making-sized hydrogen) used as a raw material is carried out, or the thermal decomposition method which makes coal-for-coke-making-sized hydrogen decompose into the bottom of high temperature. Therefore, in the case of fullerene manufacture, since a multi-annular aromatic compound and a carbon system macromolecule component are also produced in coincidence, this soot-like matter content air current turns into an air current (soot-like matter content air current) containing fullerene, a multi-annular aromatic compound, and the soot-like matter containing a carbon system macromolecule component.

[0026] Since the temperature field where the maximum temperature in a fullerene composition region is low temperature comparatively compared with about 1700 degrees C and other approaches, and fullerene exists in the state of a gaseous phase, and carbon system macromolecule components other than fullerene which occupy many of especially soot-like matter exist in the state of a solid-state tends to specify a combustion method toward mass production method of fullerene and it can separate fullerene efficiently, it is desirable. What is necessary is it to be common to carry out under reduced pressure to atmospheric pressure as a flow and pressure requirement, and just to choose whenever [reduced pressure] suitably, when manufacturing fullerene with a combustion method. As a concrete flow and pressure requirement, 1330-13300Pa (10 - 100Torr) is desirable, and further 3990-6650Pa (30 - 50Torr)

is desirable. Although what is necessary is just to choose suitably as temperature conditions according to the flow and pressure requirement which carried out point **, 800–2500 degrees C is desirable especially, and it is especially desirable that it is 1200–1600 degrees C further 1000–2000 degrees C.

[0027] As a hydrocarbon used as the raw material of fullerene, the aromatic hydrocarbon of the carbon numbers 6–20, such as benzene, toluene, a xylene, naphthalene, a methylnaphthalene, an anthracene, and a phenanthrene, is used suitably. Moreover, as coal-for-coke-making-ized hydrogen, it may use together to these aromatic hydrocarbon, and aliphatic hydrocarbon, such as a hexane, a heptane, and an octane, may be used.

[0028] The coal-for-coke-making-ized hydrogen used in a combustion method acts on coincidence also as a heat source. That is, it is thought that this coal-for-coke-making-ized hydrogen generates the carbon unit for forming a fullerene frame by carrying out the dehydrogenation of the coal-for-coke-making-ized hydrogen while raising the temperature which reacts with oxygen, generates heat and becomes generable [fullerene]. Carbon units gather on a fixed pressure and temperature conditions, and form fullerene.

[0029] Moreover, as for the ratio of a fuel required [although what is necessary is just to also choose the rate of the fuel in combustion, and air suitably] in order that an introductory air content may carry out stoichiometric combustion, and the fuel to throw in, in the volume ratio in ordinary temperature and ordinary pressure, it is usually desirable that it is 1:2–1:4, and further 1:2 to 1:2.5. As amount of the oxygen used used in a combustion method, although it changes a little also with classes of coal-for-coke-making-ized hydrogen, when toluene is used, for example as coal-for-coke-making-ized hydrogen, a 0.5 to 9 time mol is desirable to toluene, and an one to 5 time mol is still more desirable. Inert gas may be made to exist in the system of reaction in a combustion method to fullerene in addition to oxygen. In this invention, inert gas means fullerene and the gas which does not react substantially. As a class of inert gas, helium, neon, an argon, nitrogen, and such mixture are mentioned.

[0030] In the soot-like matter obtained by the combustion method, fullerene and multi-annular aromatic hydrocarbon are contained. The remainders other than these are hydrocarbons, carbon black, etc. of a giant molecule which usually have some hydrogen atom by making carbon graphite structure with graphite structure into a frame (carbon system giant-molecule component). It is desirable that fullerene is contained in the soot-like matter obtained at a process (1) 5% of the weight or more, and especially the thing included especially 15% or more is desirable 10 more% or more. Moreover, although there will be no limit in a carbon number if the fullerene manufactured by this invention has fullerene structure, it is usually the fullerene of carbon numbers 60–84, and it is desirable that the percentage of C60 and C70 is 50% or more in [all] fullerene especially, and it is desirable that it is especially 80% or more 70 more% or more.

[0031] The soot-like matter content air current acquired at a process (1) has a loose rate, and usually has the temperature of 300 degrees C or more. This air current may be offered to the following process (2) as it is, the inert gas style of point ** is supplied, for example, an air speed may be raised, and the effectiveness in a process (2) may be raised, and this soot-like matter content air current may be heated if needed. Under the present circumstances, as for the temperature of a soot-like matter content air current, it is [that what is necessary is just beyond the temperature in which fullerene and/or a multi-annular aromatic compound can exist with a gas] desirable that it is specifically 400 degrees C or more. Moreover, when the temperature of a soot-like matter content air current is too high, the load to a fullerene manufacturing installation is large, and since the increment in the effectiveness of balancing a temperature rise decreases, it is desirable also in 2000 degrees C or less that it is 1500 degrees C or less. In addition, the multi-annular aromatic compound in this invention has at least one or more rings, and shows the aromatic compound which two or more rings have condensed. Naphthalene, an anthracene, a phenanthrene, a PENZO pyrene, etc. are mentioned as an example of a multi-annular aromatic compound.

[0032] (Process 2) A process (2) separates fullerene and/or a multi-annular aromatic compound from the soot-like matter content air current containing the fullerene, multi-annular aromatic

compound, and carbon system macromolecule component which were obtained at the process (1) of point ** by the gaseous state. As a method of separation, the following (processes 2-1) and (a process 2-2) are mentioned.

(Process 2-1) A multi-annular aromatic compound is first separated from a soot-like matter content air current by the gaseous state. Next, the method of separating fullerene from the mixture of fullerene and a carbon system macromolecule component.

(Process 2-2) How to separate the multi-annular aromatic compound of the fullerene of a gaseous state, and a gaseous state first with other matter contained in soot-like matter, such as a carbon system macromolecule component, and then to separate fullerene from the mixture of fullerene and a multi-annular aromatic compound. These processes are explained.

[0033] (Process 2-1) This process separates a multi-annular aromatic compound from a soot-like matter content air current by the gaseous state. What is necessary is to consider economical efficiency and just to carry out on the optimal conditions as separation conditions in this case, that what is necessary is just to choose the temperature of arbitration, and a flow and pressure requirement suitably, if a multi-annular aromatic compound can exist by the gaseous state. generally 100 to 2×10^5 Pa of a pressure is desirable, and it is further 1000 to 1.4×10^5 Pa — it is desirable. In ordinary pressure, there is a merit to which equipment becomes easy, and since a multi-annular aromatic compound exists by the gaseous state even if separation condition temperature is low, it can dissociate, and there is an economical merit under reduced pressure.

[0034] Moreover, although separation condition temperature is based also on a pressure, it is 600 or less [100 degrees C or more] preferably. What is necessary is just to choose separation condition temperature suitably, since the temperature in which a multi-annular aromatic compound can exist by the gaseous state naturally changes with pressures. When separation condition temperature is ordinary pressure, 200 more degrees C or more 600 degrees C or less are desirable, and 300 degrees C or more especially 550 degrees C or less are desirable. If temperature is too low, multi-annular aromatic hydrocarbon may deposit as a solid-state, since fullerene will also be in a gaseous state if temperature is too high conversely, it will dissociate with a multi-annular aromatic compound, and the recovery of fullerene may fall.

[0035] Although the soot-like matter content air current acquired at the process (1) may be used as it is on the occasion of separation of a multi-annular aromatic compound, inert gas like point ** may be added further. In order to avoid the reaction of these inert gas and fullerene, it is desirable for inert gas to permute the inside of a decollator substantially, and to separate a multi-annular aromatic compound under inert gas circulation. It is desirable to make the oxygen content in the gas in a decollator below into 10 volume % especially, and also considering especially as one volume % is desirable below 5 volume %. If there are many oxygen contents, the oxide of fullerene may generate and the yield of fullerene may fall.

[0036] In the case of ***** of a multi-annular aromatic compound, it is desirable that the sum total with the amount of inert gas carried in from a process (1) as an amount of circulation of inert gas to 1g of soot-like matter containing fullerene and multi-annular aromatic hydrocarbon is 1 - 10000 ml/min, and it is desirable that they are further 5 - 5000 ml/min. Circulation of inert gas may be continuous or may be intermittent. What is necessary is just to collect the multi-annular aromatic compounds separated with the decollator as a solid multi-annular aromatic compound by being accompanied to an air current, for example, lowering temperature with deposit equipment. This deposit equipment may be formed in the same equipment as a decollator, or may be formed as another equipment. Any of a batch type or continuous system are sufficient as recovery of this multi-annular aromatic compound. As it is, as a solid-state, it may dissolve in a solvent and recovery of the depositing multi-annular aromatic compound may be collected.

[0037] The technique of making it solidifying and collecting well-known gaseous state matter conventionally should just be used for a deposit of a multi-annular aromatic compound and the approach of recovery. For example, the gas which contains the multi-annular aromatic

compound of a gaseous state in the cooled rotating drum is contacted, a multi-annular aromatic compound is deposited, and the approach of carrying out scraping recovery with a scraper intermittently or continuously is mentioned. moreover, it may dissolve with an organic solvent, and the multi-annular aromatic hydrocarbon which boiled the rotating drum and adhered may be removed and collected. moreover, gas is cooled by blowing the gas which is made to pass the multi-annular aromatic compound of a gaseous state through the spray zone of water or an organic solvent, or includes multi-annular aromatic series combination of a gaseous state as an option into water or an organic solvent, and the approach of carrying out deposit (or it dissolving in organic solvent -- making) recovery of the multi-annular aromatic compound is mentioned. Under the present circumstances, what is necessary is for an organic solvent just to extract a multi-annular aromatic compound further, when a multi-annular aromatic compound is deposited underwater and collected underwater.

[0038] After separating a multi-annular aromatic compound at a process (2-1), fullerene is separated from the mixture of fullerene and a carbon system macromolecule component. Although especially a limit does not have a method of separating fullerene and a carbon system macromolecule component, the soot-like mixture which contains fullerene and a carbon system macromolecule component, for example is mixed with an extracting solvent, and the approach (process 2-1-1) of obtaining the extract which fullerene dissolved, and the approach (process 2-1-2) of heating the soot-like mixture containing fullerene and a carbon system macromolecule component under existence of inert gas etc., and carrying out sublimation separation of the fullerene are mentioned.

[0039] (Process 2-1-1) The solvent which contains aromatic hydrocarbon preferably as an extracting solvent in the case of obtaining the extract which dissolved fullerene is used. As aromatic hydrocarbon, it is the hydrocarbon compound which has at least one benzene nucleus in intramolecular. Specifically Benzene, toluene, a xylene, ethylbenzene, n-propyl benzene, Isopropylbenzene, n-butylbenzene, sec-butylbenzene, tert-butylbenzene, 1 and 2, 3-trimethyl benzene, 1 and 2, 4-trimethyl benzene, Alkyl naphthalene, such as alkylbenzenes, such as 1,3,5-trimethylbenzene, 1, 2 and 3, 4-tetramethyl benzene, 1, 2 and 3, 5-tetramethyl benzene, diethylbenzene, and a cymene, and 1-methylnaphthalene, a tetralin, etc. are mentioned. 1, 2, 3-trimethyl benzene, 1 and 2, 4-trimethyl benzene, and a tetralin are [among these] desirable.

[0040] Further, independently or while these, two or more sorts other than the aromatic hydrocarbon mentioned above as an extracting solvent may be used for organic solvents, such as aliphatic hydrocarbon and chlorinated hydrocarbon, at a rate of arbitration. As aliphatic hydrocarbon, a ring type, a non-ring type, etc. can use the aliphatic hydrocarbon of arbitration. As an example of ring type aliphatic hydrocarbon, the thing of a monocycle type and a polycyclic type is mentioned, for example, a cyclopentane, a cyclohexane, cycloheptane, cyclooctane, etc. are mentioned by the monocycle formula. Moreover, the methylcyclopentane which is the derivative of monocycle type aliphatic hydrocarbon, an ethyl cyclopentane, a methylcyclohexane, ethylcyclohexane, 1,2-dimethylcyclohexane, 1,3-dimethylcyclohexane, 1,4-dimethylcyclohexane, an isopropyl cyclohexane, n-propyl cyclohexane, t-butyl cyclohexane, n-butyl cyclohexane, an isobutyl cyclohexane, 1 and 2, a 4-trimethyl cyclohexane, 1 and 3, a 5-trimethyl cyclohexane, etc. are mentioned. A decalin etc. is mentioned as a polycyclic type. As an example of non-ring type aliphatic hydrocarbon, n pentane, n-hexane, n-heptane, n-octane, an isooctane, n-nonane, n-Deccan, n-dodecane, n tetradecane, etc. are mentioned.

[0041] As a chlorinated hydrocarbon, dichloromethane, chloroform, a carbon tetrachloride, a trichloroethylene, tetrachloroethylene, 1,2-dichloroethane, 1,1,2,2-tetrachloroethane, a chlorobenzene, a dichlorobenzene, 1-chloronaphthalene, etc. are mentioned. In addition, a with a carbon numbers of six or more ketone, with a carbon numbers of six or more ester, with a carbon numbers of six or more ether, a carbon disulfide, etc. may be used.

[0042] Since the extraction efficiency of fullerene will fall if the solubility of the fullerene in an extracting solvent is too low, it is desirable that it is $\geq 5\text{g/l.}$ or more as solubility of fullerene, and it is desirable that 10 moreg /or more is [l.] 15g/l. or more especially. Moreover, the boiling point is suitable for a 120-250-degree C thing also in 100-300 degrees C with the

ordinary temperature liquid in a industrial viewpoint to these extracting solvents.

[0043] Only the amount which can fully extract fullerene needs to be used for an extracting solvent. Usually, when the amount of 5 – 400 weight double and economical efficiency are considered to the fullerene content in soot-like mixture, it is desirable to carry out the amount extent use of 40–200 weight double. Formats, such as a batch type, semi continuous system, continuous system, or those combination, and especially equipment are not limited for an extract. In addition, although 5 – 30% of the weight of fullerene is usually contained in soot-like mixture, since it is desirable to consider as the range which mentioned above the amount of the extracting solvent used to fullerene from a viewpoint of extraction efficiency, it is desirable to analyze some soot-like matter and to measure the fullerene content in the soot-like matter in advance of extract operation.

[0044] As an extractor, a churning mixing chamber can use it suitably. In the case of an extract, there is no pressure in a container and it should just carry out especially a limit by ordinary pressure. As temperature at the time of an extract, it is usually –10–150 degrees C, is 5–80 degrees C preferably, and is 30–50 degrees C still more preferably. If it is these range, it is desirable from the field of the improvement in extraction efficiency, but since temperature dependence is small, as for extraction efficiency, it is advantageous to carry out with ordinary temperature extent in energy cost. In an extract process, if it extracts if needed further, irradiating a supersonic wave etc. at an extract, since extract time amount will become short, it is desirable.

[0045] In this way, since fullerene is dissolving in the obtained extract and the polynuclear aromatic compound is already removed at the front process (process (2-1)), the fullerene of a high grade can be obtained by distilling off the organic solvent used from the extract etc. in addition -- the slurry which remained after extracting fullerene -- the shape of an industry -- the useful carbon system macromolecule component is contained. Therefore, it is possible to carry out a non-melt a ** exception and to reuse it from this slurry. Although a ** exception is not limited, its pressure filtration is [approaches, such as filtration under reduced pressure, pressure filtration, gravity filtration, filter filtration, or those combination, and especially equipment] desirable especially.

[0046] (Process 2-1-2) The fullerene and the soot-like mixture containing a carbon system macromolecule component with which the multi-annular aromatic compound was separated can separate fullerene from soot-like mixture by the gaseous state by giving under a high temperature service rather than the temperature at the time of separating a multi-annular aromatic compound by the gaseous state. Since fullerene generally serves as a solid-state after separating a multi-annular aromatic compound, by heating soot-like mixture, fullerene is sublimated and it can dissociate.

[0047] The conditions at the time of sublimating are carried out under ordinary pressure or reduced pressure of about 5000Pa. In ordinary pressure, there is a merit to which equipment becomes easy and there is a merit to which the sublimation temperature of fullerene becomes low under reduced pressure. What is necessary is to consider economical efficiency and just to carry out on the optimal conditions. The sum total with the amount of inert gas carried in from a front process to 1g of soot-like mixture in inert gas, such as nitrogen or helium, is 1 – 10000 ml/min. It is desirable extent and that it is 5 – 5000 ml/min extent preferably. Circulation of inert gas may be continuous or may be intermittent.

[0048] If a permutation is not fully carried out, the oxide of fullerene may generate. The preheating of the inert gas at the time of sublimating may be carried out, and it does not have to carry out a preheating. As for the equipment used for sublimation, a batch type, a fixed-bed mold, a fluid bed mold, a continuation mold, etc. do not carry out especially limitation. This process separates fullerene from the air current containing soot-like mixture or this by the gaseous state. What is necessary is to consider economical efficiency and just to carry out on the optimal conditions as separation conditions in this case, that what is necessary is just to choose the temperature of arbitration, and a flow and pressure requirement suitably, if fullerene can exist by the gaseous state. As for a pressure, generally, it is desirable to carry out under

reduced pressure of about 5000Pa. In ordinary pressure, there is a merit to which equipment becomes easy, and since fullerene exists by the gaseous state even if separation condition temperature is low, it can dissociate, and there is an economical merit under reduced pressure. [0049] Moreover, it is usually especially desirable [temperature], although separation condition temperature is based also on a pressure that it is 800 degrees C - 1100 degrees C further 600-1200 degrees C 400 degrees C - 1400 degrees C. What is necessary is just to choose separation condition temperature suitably, since the temperature in which fullerene can exist by the gaseous state naturally changes with pressures. Since fullerene will not be in a gaseous state enough if temperature is too low, if yield may fall and temperature is too high conversely, when becoming disadvantageous economically and slight oxygen exists, this oxygen may react with fullerene, and may become an oxide, and the yield of fullerene may fall.

[0050] (Process 2-2) Next, the fullerene of a gaseous state and the multi-annular aromatic compound of a gaseous state are separated with other matter contained in soot-like matter, such as a carbon system macromolecule component, and how to separate fullerene is explained from the mixture of fullerene and a multi-annular aromatic compound below. Fullerene and a multi-annular aromatic compound are separable from this soot-like matter content air current by the gaseous state attaching the soot-like matter content air current which contains the fullerene and the multi-annular aromatic compound which were obtained at the process (1), and a carbon system macromolecule component at this process (2-2) under the condition of the process (2-1-2) of point **. The separation conditions in this case are the same as the process (2-1-2) of point **. Thus, generally the fullerene and the multi-annular aromatic compound of a gaseous state which were separated cool these, and make them the mixture of the shape of the shape of a solid-state, and a liquid. Subsequently, what is necessary is just to separate fullerene and a multi-annular aromatic compound by approach like the process (2-2-1) shown below, for example, a process (2-2-2), and a process (2-2-3).

[0051] Process (2-2-1)

The process which cools, uses as such shape of a solid-state and liquid mixture the fullerene and the multi-annular aromatic compound which were obtained by the gaseous state through a process (2-2), heats this mixture, and separates a multi-annular aromatic compound by the gaseous state.

Process (2-2-2)

The process which separates fullerene as a solid-state, cooling the fullerene and the multi-annular aromatic compound which were obtained by the gaseous state through the process (2-2), and making a multi-annular aromatic compound into a gaseous state.

Process (2-2-3)

The process which it cools, the fullerene and the multi-annular aromatic compound which were obtained by the gaseous state are used as such shape of a solid-state and liquid mixture through a process (2-2), and the solubility of fullerene extracts a multi-annular aromatic compound from this mixture with an organic solvent with the high solubility of a multi-annular aromatic compound low, and is separated. Each [these] process is explained.

[0052] Process (2-2-1)

In a process (2-2-1), it cools and let the fullerene and the multi-annular aromatic compound (gas) which were obtained by the gaseous state be such shape of a solid-state and liquid mixture. And this mixture is heated and fullerene and a multi-annular aromatic compound are separated. As conditions for this separation, if fullerene and a multi-annular aromatic compound are separable by the gaseous state, the temperature of arbitration and a pressure can be taken. For example, what is necessary is just to separate fullerene and a multi-annular aromatic compound by the gaseous state under the same temperature as the time of separating a multi-annular aromatic compound from the soot-like matter content air current in the process (2-1) of point ** by the gaseous state, and a flow and pressure requirement. The temperature in this case and a flow and pressure requirement consider economical efficiency, and should just carry it out on the optimal conditions. Especially, it is desirable also in this process (2-2-1) to consider as the desirable range in the same temperature as the time of separating a multi-

annular aromatic compound from the soot-like matter content air current in the process (2-1) of point ** by the gaseous state and a flow and pressure requirement.

[0053] Process (2-2-2)

In a process (2-2-2), it becomes liquid and fullerene cools the fullerene and the multi-annular aromatic compound (gas) which were obtained by the gaseous state even in the shape of a solid-state, and the condition that it is separable with the multi-annular aromatic compound of a gaseous state so that only a multi-annular aromatic compound may be in a gaseous state that is, and fullerene and a multi-annular aromatic compound are separated. as the conditions for this separation -- fullerene -- the shape of a solid-state -- or if it becomes liquid and a multi-annular aromatic compound can exist by the gaseous state, the temperature of arbitration and a pressure can be taken. for example, the bottom of the same temperature as the time of separating a multi-annular aromatic compound from the soot-like matter content air current in the process (2-1) of point ** by the gaseous state, and a flow and pressure requirement -- fullerene -- the shape of a solid-state -- or what is necessary is to suppose that it is liquid and just to separate a multi-annular aromatic compound The temperature in this case and a flow and pressure requirement consider economical efficiency, and should just carry it out on the optimal conditions. Especially, it is desirable also in this process (2-2-2) to consider as the desirable range in the same temperature as the time of separating a multi-annular aromatic compound from the soot-like matter content air current in the process (2-1) of point ** by the gaseous state and a flow and pressure requirement.

[0054] Process (2-2-3)

It is the process at which the solubility of fullerene extracts a multi-annular aromatic compound from the shape of a solid-state containing fullerene and a multi-annular aromatic compound, and liquid mixture low at this process (2-2-3), using an organic solvent with the high solubility of a multi-annular aromatic compound as an extracting solvent. this extracting solvent, i.e., the mixed solvent which contains the amides of the carbon numbers 3-5 of the carbon numbers 2-5, such as a ketones; tetrahydrofuran of the carbon numbers 3-5, such as an alcohols; acetone of the carbon numbers 1-4 of a methanol, ethanol, propanol, ethylene glycol, a glycerol, etc. and a methyl ethyl ketone, diethylether, and dioxane, such as ether; N.N-dimethylformamide, and these as an example of a solvent (a poor solvent may be called hereafter) with the low solubility of fullerene, is mentioned. Alcohols are desirable, especially, a with a carbon number of three or less thing is desirable, and especially 2-propanol (isopropyl alcohol) is [among these] desirable.

[0055] As solubility of the fullerene C60 of these poor solvents, it is desirable that it is 1g/l. or less, and it is desirable that 100 moremg /or less is [l.] especially 50 or less mg/L. Although it may be directly contacted into the shape of a solid-state of point **, and liquid mixture, generally, a poor solvent dissolves in the organic solvent which the both sides of fullerene and a multi-annular aromatic compound show high solubility, once uses this mixture as a solution, adds a poor solvent to this and deposits fullerene.

[0056] As for the amount of the poor solvent used in this case, it is desirable that it is amount extent of 1 - 30 weight double to the amount of organic solvents used for this solution adjustment also in the amount of 0.1 - 50 weight double. If there are few amounts of a poor solvent, the amount of deposits of fullerene will decrease and recoverable fullerene will decrease. If many [too], the container capacity which performs these actuation will become large, and a loss will occur economically. As temperature which mixes a poor solvent, it is -20-150 degrees C, and it is usually desirable especially that it is 30-60 degrees C further 10-80 degrees C -10-100 degrees C. What is necessary is for filtration etc. just to recover the fullerene which deposited by mixing a poor solvent. On the other hand, since most multi-annular aromatic hydrocarbon is dissolved into a solvent, without depositing, the solution after removing fullerene can collect multi-annular aromatic compounds as a solid-state by distilling off the solvent etc.

[0057]

[Example] Although an example is shown below and this invention is explained to it still more

concretely, unless the summary of this invention is exceeded, this invention is not limited to the following examples.

[0058]

[Example 1] 3.8mg of soot-like matter obtained by the combustion method by using toluene as a raw material was heated from the room temperature to 1150 degrees C in 20-degree-C per minute in desiccation nitrogen gas 100 cc/min using the apparatus for thermogravimetry (TG-DTA6300 by SEIKO CORP.), and change of weight was measured. The obtained result is shown in drawing 2. In drawing 2, as for an axis of abscissa, a right longitudinal shaft shows [the rate / as opposed to the weight of 3.8mg in a left-vertical shaft / of loss in quantity] whenever [stoving temperature] for the rate of change of the rate of a raw material. A passage clear from the graph which was shown in drawing 2 and which shows weight reduction, and the graph which shows weight rate of change, if temperature becomes 100 degrees C or more, weight will decrease gradually, and it turns out that weight reduction is accelerated from near 400 degree C. And in an elevated-temperature field 500 degrees C or more, the weight of the soot-like matter decreases rapidly. This is that a lot of fullerene in the soot-like matter will sublime if it takes into consideration that the sublimation temperature of fullerene is 400-800 degrees C, and it turns out that the rapid decrease of weight of the soot-like matter arose.

[0059] . Furthermore, qualitative analysis of the gas constituents generated when the soot-like matter was heated was performed on condition that the following using the quadrupole form mass spectroscope (two to JEOL auto mass AM15 mold). A result is shown in drawing 3.

A measuring method : The EI method furnace section temperature : 290 degrees C Transfer tube temperature : 285-degree-CGC oven temperature : 285-degree-C interface temperature : 285-degree-C ionization chamber temperature : 260-degree-C photograph mull electrical potential difference : 450V ionizing voltage : 70eV ionization current : 300microA mass range: 10 - 400amu scan speed : 1000msec [0060] In drawing 3, as for an axis of abscissa, an axis of ordinate shows whenever [stoving temperature] for the relative intensity of an ion spectrum. From drawing 3, the peak which shows existence of multi-annular aromatic compounds, such as aromatic compounds, such as benzene (molecular weight 78), toluene (molecular weight 92), and a xylene (molecular weight 106), and naphthalene (molecular weight 128), an anthracene (molecular weight 178), was checked in the temperature requirement lower than the sublimation temperature of fullerene in the gas which heated the soot-like matter and occurred. As for this, a multi-annular aromatic compound also shows that it is separable by the gaseous state as well as aromatic compounds, such as benzene. Fullerene and/or a multi-annular aromatic compound are separable from the soot-like matter which contains the fullerene and the multi-annular aromatic compound which were obtained by the combustion method, and a carbon system macromolecule component according to the above example by the gaseous state. Therefore, from the air current containing such soot-like matter, it is possible to separate fullerene and/or a multi-annular aromatic compound by the gaseous state continuously, and fullerene is continuously understood are disengageable from the air current containing this soot-like matter.

[0061]

[Effect of the Invention] According to this invention explained above, it becomes possible to be able to separate manufacture of fullerene, and fullerene and other soot components continuously within one equipment, and to produce a lot of fullerene continuously. When the fullerene which attracts attention since various as the exotic material which bears the next generation, and new materials is manufactured on a scale of industrial, a lot of soot-like matter and fullerene which are generated can be efficiently separated and collected to coincidence.

[Translation done.]

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] The whole example outline sectional view of the fullerene composition decollator concerning this invention.

[Drawing 2] Drawing having shown the weight reduction at the time of heating the soot-like matter.

[Drawing 3] Drawing showing the qualitative-analysis result of the gas which occurred when the soot-like matter was heated.

[Description of Notations]

1: Fullerene generation equipment

2: Burner

3: Decollator

4: Decompression device

5: Deposit equipment

6: Recovery system

[Translation done.]

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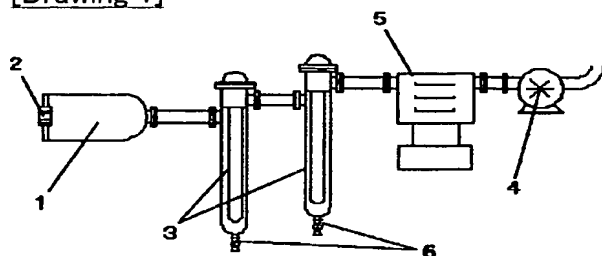
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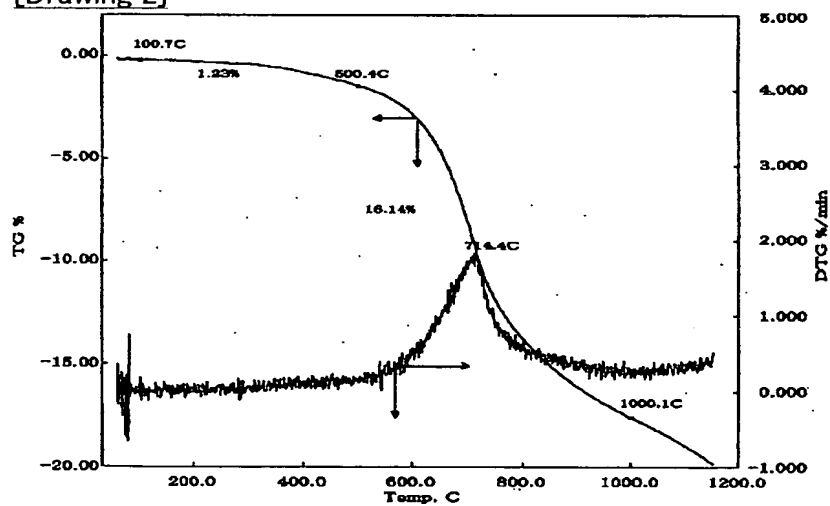
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DRAWINGS

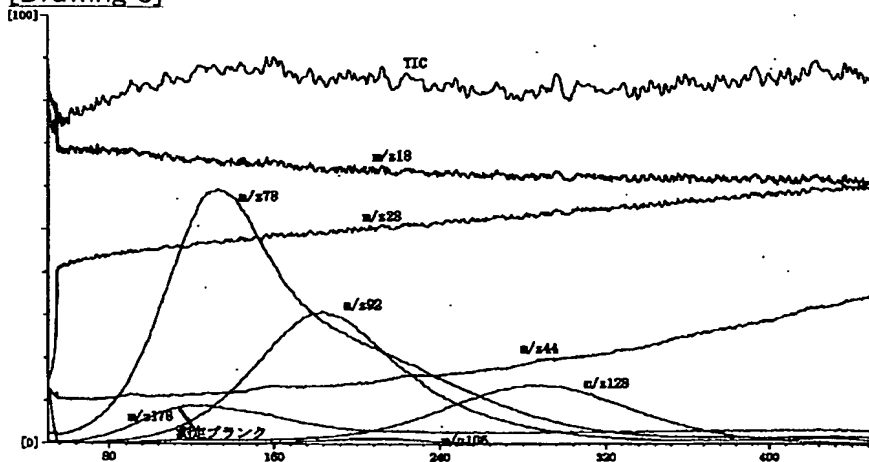
[Drawing 1]



[Drawing 2]



[Drawing 3]



[Translation done.]